

# Identifying New Biogeochemistry Science Opportunities at DOE Sites

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## **1.) Describe major remediation activities underway at your site.**

The Hanford site is divided into two major regions where contamination exists. The first is the Hanford plateau that exists in the site center. Processing of irradiated nuclear fuels and Pu extraction occurred in the central plateau where ca 150' of unsaturated vadose zone overrides an unconfined aquifer ranging in depth from 100' to 10' thick. Tank farms, specific retention basins, cribs, and many trenches exist in the central plateau where over 10 distinct groundwater plumes exist. Two forms of remediation have been applied in the central plateau, vapor extraction for CCl<sub>4</sub> and pump-and treat for U(VI)/Tc(VII). Pumping operations were effective for Tc(VII) but not U(VI). Corrective measures (barriers) as soon to be applied to minimize seasonal infiltration in the tank farms.

The second region is the Columbia River corridor that cuts through the entire site. It is characterized by more shallow groundwater (~20-40'), and aquifer connectivity with the Columbia River. Contaminants released to groundwaters in the central plateau move to and discharge to the Columbia River through the River Corridor. Eight reactors, many infiltration trenches, solid waste burial grounds, and a major process-pond complex exist in the river corridor. Contaminated sites are currently being remediated by excavation to a depth of 15'. Plumes of Cr (multiple), <sup>90</sup>Sr, and U originate from contamination in the river corridor. Pump and treat has been applied in both the <sup>90</sup>Sr and Cr plumes, but was found ineffective for the former. A 100 m+ reductive barrier has been placed in the near-shore region to prevent Cr discharge to the Columbia River. The barrier employs dithionite as a sediment reductant and has been partially effective. A form of biologically driven, in-situ apatite precipitation has been proposed for the <sup>90</sup>Sr plume, but there appear to be problems in deployment.

## **2.) Who is performing the site remediation.**

Flour-Hanford is in charge of groundwater remediation site-wide. Bechtel is in charge of river corridor source term removal by excavation and long term storage in ERDF in central Hanford. The CH2M-Hill Hanford group oversees tank farm characterization, waste retrieval, and tank farm closure and vadose zone remediation.

## **3.) What are the primary risk driving contaminants?**

Mobiles: <sup>99</sup>Tc ~ <sup>129</sup>I ~ CCl<sub>4</sub> > Cr

Semi-mobile: U(VI) > <sup>90</sup>Sr

Less mobile: <sup>137</sup>Cs

Immobilized: Pu

Honorable mention?

#### **4.) Major subsurface contamination problems?**

The migration of mobile  $^{99}\text{Tc}$  from multiple high level waste tank leaks (SX-018, T-106) and the BC cribs in the central plateau out to the Columbia River.

The migration of  $\text{CCl}_4$  as a DNAPL below the Z-cribs to the base of the unconfined aquifer and its through migration as a dissolved solute to the Columbia River.

The continued breakthrough of multiple vadose zone U plumes in the central plateau and other locations to groundwater, and their retarded transport to the Columbia River.

#### **5.) What are the primary problems facing closure and restoration of Hanford?**

Difficulty in characterizing the vertical and spatial distribution of contaminants.

Dealing with large reservoirs of potentially mobile constituents in the deep vadose zone.

The high level waste tanks and their in-tank and in-ground residuals.

Effective, acceptable, and economically viable remedial techniques.

Scientific information on subsurface geochemical, hydrologic, and microbiologic processes.

Staying power of US taxpayers given other societal concerns.

#### **6.) Two critical remedial needs.**

$^{99}\text{Tc}$  in deep vadose zone sediments and deep groundwaters of the Hanford central plateau.

$\text{CCl}_4$  in deep vadose zone sediments and deep groundwaters of the Hanford central plateau.

#### **7.) Balance between engineered solutions and natural attenuation.**

DOE/Hanford is only now entering the phase of remedial decision making that is accomplished through the Tri-Party Agreement with US EPA and the Washington state DOE as participants.

The first sites being considered are contaminant plumes discharging directly to the Columbia River (100 N –  $^{90}\text{Sr}$ , and 300 A – U). DOE will push natural attenuation for all but the most contaminated locations. The regulators will push back and will succeed in forcing DOE to implement engineered solutions for key sites. These

battles are forthcoming, with the first to occur within the next 3-5 years. It will probably be at least 10 years before the first decision is made for the least contaminated of the tank farms. There are schedules in place for when these decisions are mandated to occur within context of the TPA.

In the end, I estimate that MNA will be applied to 70% of the sites and engineered solutions to 30%. Most of the engineered solutions will be barriers designed to eliminate the small amount of recharge that occurs in Hanford's semi-arid environment.

## **8.) Biogeochemical Processes at Hanford.**

There is surprisingly little known about subsurface microbiology at the Hanford Site. Almost all of this information has originated from OBER supported research: i.) deep subsurface coring and microbial characterization at the Yakima barricade and Savage Island in the early 1990's, ii.) a short-term NABIR-EM linkage project (300 A near-river aquifer), iii.) NABIR-LBNL field manipulation at 100 D., and iv.) RACS-funded microbiological characterization of the extreme (radiation, pH, temp) of the SX-108 borehole.

Generally, the deep vadose zone is found to have few culturable organisms which are attributed to its very low moisture content, coarse texture, and limited recharge below the rooting zone. Artificial recharge increases subsurface populations by transporting soil organisms to depth and providing adequate moisture for nutrient access and metabolism.

Very little information exists on the unconfined aquifer that underlies almost all of Hanford, and that is the primary receiving water for contaminants from the Hanford vadose zone. This lack of knowledge stems from the fact that it is very difficult to retrieve aseptic aquifer sediments for study that have not been compromised by the drilling and sampling operation. Most significant is the abrasion and crushing of basalt clasts that expose highly reducing ferrous-silicates surfaces that react with water and generate hydrogen. Another complicating issue is that groundwater in the unconfined aquifer is very low in organic carbon, and only support a limited microbiologic community.

Recent studies by Jim Fredrickson and colleagues using near-shore aquifer sediments from the 300 A show distinctly larger bacterial populations that may drive significant biogeochemical processes. Organisms, for example, were isolated from these sediments that are capable of Tc(VII), U(VI), and Fe(III) (Pena bacillus). Additional studies are proposed to characterize these potentially active biogeochemical zones. Cores from deeper, more fine-grained sediments at this location show signs of more active biologic Fe(III) reduction.

## **9.) Are there biogeochemical issues associated with long-term stewardship?**

One would certainly think so, but maybe this is the \$99 question. It may be that these are very slow and difficult to study processes occurring at native rates controlled by water infiltration, and fluxes of carbon and electron donors. Here are some potential issues:

- Could bacterially driven dissolution/precipitation processes lead to residual contaminant structural incorporation or sequestration. (carbonates, aluminosilicates)
- Pu and Am will likely be left in place because of their perceived and measured immobility under short-term study. Could they be mobilized by future microbiological processes involving biogenic complexants?
- Could dissimilatory N reduction of large amounts nitrate/nitrite lead to mobilization of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ , or other adsorbed cations by biogenic ammonium/ion exchange displacement.
- Are there biological processes that could influence  $^{129}\text{I}$ , e.g., volatilization or other speciation changes.
- Could biologic processes impact the long-term stability of remedial schemes involving phosphates, such as apatite or autunite precipitation?
- Could changing groundwater levels or gradients at Hanford lead to slower groundwater velocities and lower DO concentrations? Note massive amounts of artificial recharge during Hanford site operations and currently ongoing changes including wells going dry and plume directional reversals.